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6. AUTHOR(S)
Lawrence A. Crum

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Applied Physics Laboratory
University of Washington
1013 NE 40th St.
Seattle, WA 98105-6698

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9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
Office of Naval Research
800 N. Quincy St.
Arlington, VA 22217-5660
    Attn: Dr. Peter P. Schmidt, Code 331

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13. ABSTRACT (Maximum 200 words)
The objectives for this project were to better understand sonoluminescence conditions, expand the parameter space for sonoluminescence, enhance the cavitation collapse sequence, and implement enhanced-cavitation generators to search for tritium production. Modeling studies were performed to determine additional paths to unknown stable parameter spaces. Experiments were performed to determine methods and mechanisms for enhancing cavitation collapse. PZT array and spark-gap lithotripter-based systems were designed, built and tested. The spark-gap lithotriper worked as planned, but the count rate was too slow for practical applications. The PZT array system worked, but final testing was not completed because funding was exhausted.

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Final Report

GRANT NUMBER: N00014-99-1-0793
PROJECT: Energy Focusing in high-amplitude sound fields

OBJECTIVES

The objectives for this project were to better understand sonoluminescence conditions, expand the parameter space for sonoluminescence, enhance the cavitation collapse sequence, and implement cavitation generators to search for tritium production. In particular, the goals of this research were:

(1) Understanding sonoluminescence: Towards a better understanding of sonoluminescence, we designed and built a sonoluminescence system to test for X-ray emissions.

(2) Expanding the sonoluminescence parameter space: We fabricated low and high frequency levitation systems, and designed and built a levitation system for shock wave studies. We also examined different liquids. A theoretical mapping to additional parameter spaces was undertaken.

(3) Enhancing cavitation collapse: To enhance cavitation collapse we designed and built two cavitation generators, a shock wave generator, and a low frequency array cavitation generator.

(4) Tritium detection: Experiments were conducted on systems designed and built to test for tritium production.

RESULTS

The results for each objective are summarized below. Please see details in the annual project reports.

(1) Understanding sonoluminescence:

(1a) A high-risk, high-payoff experiment was conceived and implemented to measure X-ray emissions from a sonoluminescence bubble. The experiment was conducted by a postgraduate student and collaborators from the University of Washington's Physics Department. Experiments were conducted at the APS, Argonne National Laboratory. The experiments proved inconclusive.

(1b) Understanding the mechanisms of sonoluminescence is required for controlling and enhancing the emission from sonoluminescing bubbles. The work validated the dissociation hypothesis, using many combinations of gases, and indicated how this hypothesis works in interpreting multiple frequency drive. The hypothesis breaks down for concentration of air greater than about 50%, when the chemical equilibrium established between the liquid and bubble is such that not all of the nitrogen can exit the bubble, even under the conditions of a plasma (Ketterling 1999; Ketterling and Apfel 2000).

(2) Expanding the sonoluminescence parameter space:

(2a) A theoretical mapping of additional parameter spaces was conducted by the subcontractor at JHU. The objective was to determine the theoretical constraints for increasing the expansion ratio using multiple-frequency drives. Initial stable configurations of multiple harmonic drives were determined. A typical optimization for a first and second harmonic drive system compared to a single frequency drive implies that the optimized multi-mode driving system can generate an order of magnitude increase in the bubble's expansion ratio (R_{max}/R_0).

The second approach utilized an active control strategy. One may envisage that, in an experiment, the bubble distortion is continuously monitored (e.g., by a cross-correlation of light scattered in two different directions), and this information is used to modulate the sound field in such a way as to maintain the bubble spherically stable. We have tried two different controls, one based on the bubble velocity, the other based solely on the amplitude of the shape mode instability (the prolate-oblate shape mode).

Implementation of the control is effective in removing the shape instability, but so far our conclusions imply that only a very modest increase in the allowable acoustic amplitude can be achieved in this way. Thus, the provisional conclusion is that the multi-frequency approach described first is far more effective in addition to being easier to implement in actual experimental practice. We are thus focusing our efforts in this direction.

(2b) Low and high-frequency levitators were designed and built and tested to expand the parameter space for sonoluminescence. In particular, we hypothesized that lower frequencies would generate more intense collapse. During our investigation, we discovered that vapor trapping within the bubble interior limits the degree to which a bubble's collapse can be violent. This limitation motivates our need for using low vapor pressure liquids.

(2c) A levitation system was designed and built to examine how shock waves force a bubble into growth and collapse. The experiment proved very successful, and was used subsequently in our design of a shock wave generator for future experiments (Matula, Hilmo et al. 2002; Matula, Hilmo et al. 2002).

(3) Enhancing cavitation collapse:

We designed and built alternative systems for creating violent cavitation collapse. These systems included (1) an electrohydraulic shock-wave device, and (2) a Tone-Burst Low-frequency PZT array device (Matula and Hilmo 2003).

The electrohydraulic shock-wave device can generate up to about negative 100 Atm, and can therefore generate much greater cavitation collapse conditions than a standing wave system. Our system worked as planned (see technical reports for years 1 and 2). However, because of the spark discharge, the system could only operate at about 4 Hz maximum, resulting in slow pulse repetition frequency (PRF). In addition, we did not have access to a pulsed neutron source, so that not every spark discharge generated a

positive cavitation event. Taken together, this system was considered to be too inefficient to generate fusionable cavitation with a small asynchronous neutron source. An improved lithotripsy system would greatly help.

The concept of an improved shock wave generator led us to build a small PZT array system. It was tested and modified for use with deuterated acetone. Excellent results were obtained from this system (see technical reports for years 2 and 3). Unfortunately, final testing was not completed because funding expired. During the last week of funding, we nevertheless utilized the PZT system in a -20^0C freezer, using an asynchronous 'hot' neutron source. The experiments were conducted at the Nuclear Physics Lab on the University of Washington campus. The system was left running for the entire week, but because of time constraints and lack of funds, no monitoring was performed during this crucial test. After the week was over, the liquid was transferred into small vials and hand-carried to the tritium testing facility at the University of Washington (see details in year 3 technical report). Several days later the results were returned. No tritium was observed above background. Unfortunately, because of a lack of monitoring and funds, we could not verify these results. Future funding would help resolve some of the issues described here and in the previous technical reports, and allow a more thorough investigation into the possibility of nuclear fusion from cavitation bubbles.

(4) Tritium detection:

Because we did not have access to a neutron detector capable of being used in the shock wave and array systems, we concentrated on tritium measurements. After an experiment was run, samples were sent to the UW Medical Center for tritium analysis. Tritium measurements were performed as a thyroid bioassay: The contents of each 1-ml vial was mixed with a solution that resulted in luminescence. The count rate, in counts per minute (CPM) were measured over multiple days, to account for any anomalous readings. The same procedure was also used on a control vial of liquid (background). The final results were calculated as $MDC = 4.65\sqrt{Background / Time}$. Statistical measures of tritium would be greater than MDC.

REFERENCES

J. A. Ketterling (1999). An experimental validation of the dissociation hypothesis for sonoluminescence and an extension to the analysis of multiple frequency drives, Yale University.

J. A. Ketterling and R. E. Apfel (2000). "Shape and extinction thresholds in sonoluminescence parameter space." J. Acoust. Soc. Am. 107(3): L13-L18.

T. J. Matula and P. R. Hilmo (2003). Alternative ways to generate hot bubbles. Meeting of the Acoustical Society of America, Nashville, TN.

T. J. Matula, P. R. Hilmo, et al. (2002). "In vitro sonoluminescence and sonochemistry studies with an electrohydraulic shock-wave lithotripter." Ultrasound in Med. & Biol. 28(9): 1199-1207.

T. J. Matula, P. R. Hilmo, et al. (2002). "Radial response of individual bubbles subjected to shock wave lithotripsy pulses in vitro." Phys. Fluids 104: 913-921.

OBJECTIVE

The objective of this project has three basic goals. (1) To determine how sonoluminescence can be scaled to higher energy concentrations, (2) to increase the parameter space in which sonoluminescence currently exists, and (3) to learn more about the light emission mechanism(s). Our FY01 milestones included:

(1) To measure the light emission from a lithotripter bubble in low-vapor fluids.

(2) To build a system to measure the plasma state within the bubble.

(3) To test the apparatus for low-frequency systems.

(4) To finish the optimization for multi-frequency drives.

The goals of this research are to better understand the mechanism of sonoluminescence, and provide vectors for enhancing the light output.

APPROACH

Our approach to a better understanding of sonoluminescence involved exploring new parameter spaces for bubble emissions, and trying to probe the bubble interior using x-rays. Each objective was designed to probe and/or enhance the light emission from a sonoluminescing bubble. For objective (1), we utilized our current research lithotripter to examine how a bubble responds to dramatic pressure amplitudes. The lithotripter generates peak positive pressures of up to 1,000 atmospheres, and peak negative pressure of around 100 atmospheres. Figure 1 illustrates our spark-generated lithotripter and a typical measured spatial waveform from our lithotripter.

Figure 1. A diagram of the electrohydraulic shock wave lithotripter (SWL). A spark discharge located at the first focus of an ellipsoidal reflector is reflected by the ellipsoid and focused at the second focus, F2. Note that there also exists a direct diverging wave, originating from the spark gap, and passing through F2. Although small, the effect of this wave can be observed. The spatial profile of a measured shock wave is also shown.
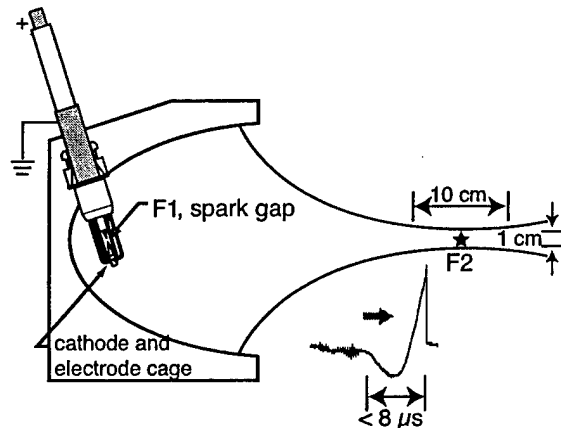
The large pressure waveforms are obtained by generating an underwater explosion using a spark discharge (15-20 kV). The explosive acoustic pulse is then reflected by an ellipsoidal reflector, focusing the waveform at the second focus of the ellipsoid, F2. A preexisting bubble located at F2 will first experience the positive pulse, followed by the long negative tail.

Because normal single-bubble sonoluminescence (SBSL) only utilizes approximately 1 atmosphere (above ambient), we expect that the bubble dynamics from a lithotripter pulse to be much more energetic than from SBSL bubbles, possibly leading to much more interesting states of matter within the compressed bubble interior.

We used the lithotripter "as-is", and built a light-scattering system to measure the bubble dynamics. We also designed and built a small enclosure to mount a photon detector (PMT) to record the light emission. The small enclosure also allowed us to fill the volume with various liquids (see

Figure 2). Bubble dynamics measurements, light intensity measurements, and acoustic emission measurements were used to probe the bubbles. Besides water, we also studied bubble effects from silicone oil, dodecane, and acetone.

Figure 2. An enclosure was designed and built to house different fluids, as well as the detection apparatus. Furthermore, we were able to mount an SBSL levitation cell to the apparatus so that single bubbles could be levitated and placed in the path of the lithotripter pulse. Single bubble levitation required us to surround most of the cell with an air gap so that a standing wave profile could be reinforced. Only two small sections of the cell were coupled directly to the outside water tank, so that the lithotripter pulse could pass through relatively unimpeded. Because of the total enclosure, a heating wire was used to generate bubbles, which were then pulled to the chamber's pressure antinode. The light emission from the bubble(s) were focused before entering the PMT detector. Calibration of the system was performed using normal SBSL under the same geometry.
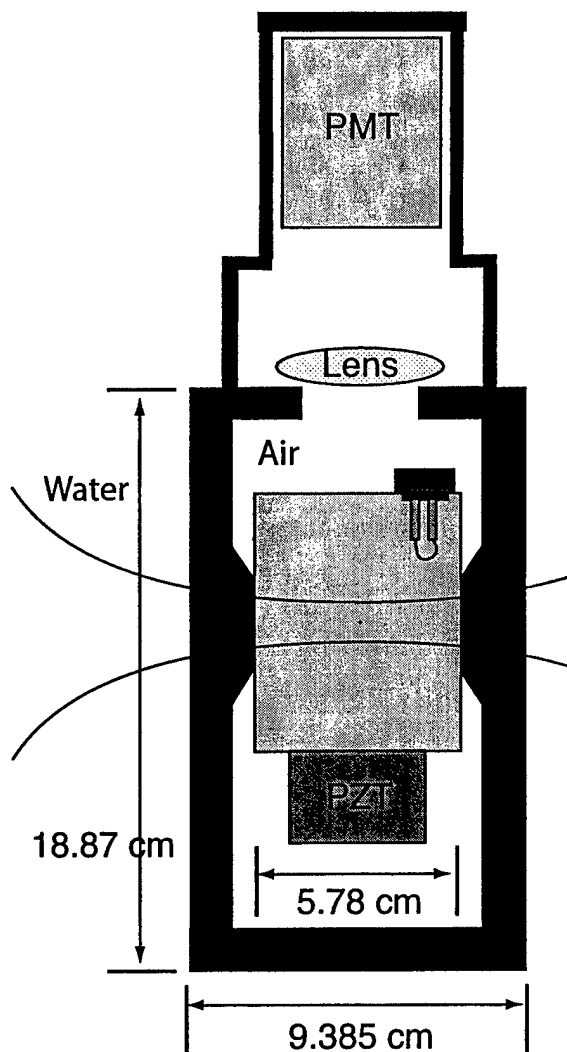
For objective (2), we designed and built a system to probe the bubble interior using x-rays. By using the Argonne National Labs synchrotron source, we were in a position to observe how x-rays interact with the assumed plasma state within the bubble. Assuming that a plasma exists within the bubble interior, we expect that the x-ray beam should alter

the plasma state by stripping k-shell electrons from their bound state, which would change the plasma conditions, and alter the light emission characteristics. For this objective we hired a postdoc with extensive knowledge of nuclear physics, and a background in SBSL. The postdoc designed and built an apparatus that could be inserted into one of the housing units (a DOE sponsored unit that is run by the physics department at the University of Washington). The bubble levitation chamber was mounted on a 3-d computer-controlled positioning system so that remote manipulation of the apparatus could be done outside the restricted housing unit when the x-ray beam was operating. A light-tight enclosure surrounded the levitation chamber because of the stray light existing in the room. X-ray intensities and beam profile were manipulated using the in-house system. Prior to the experiments, calculations were performed to determine the x-ray penetration path and appropriate gas for the study.

Objective (3) was designed to increase the collapse strength of a sonoluminescing bubble, by forcing it to grow over an extended period of time using lower frequencies. By decreasing the frequency, the expansion time increases, resulting in a larger bubble prior to collapse. For this objective, we designed and built several low-frequency levitation cells, operating between 5-7.5 kHz. Once stable sonoluminescence was achieved, light-intensity measurements were conducted to compare with standard SBSL emission intensities (from cells operating in the 20-30 kHz regime). Most of the work for this objective was accomplished during Year 1, and will not be emphasized in this Year-2 report.

Objective (4) was based on the notion that the inclusion of higher-order standing wave modes could enhance the growth, or collapse strength of the bubble (Holzfuss, Ruggeberg et al. 1998). Our goals were to determine how the higher order modes affected the bubble oscillations, optimize the higher-order modes through calculations, and compare those with experiment. Most of the work for this objective was accomplished in Year 1, and will not be emphasized in this Year-2 report.

PROGRESS

Based on new evidence that low frequency drives are negatively impacted by the presence of water vapor (Storey and Szeri 2000; Toegel, Gompf et al. 2000), and finding ourselves that low-frequency systems do not greatly enhance the light emission, we cut short our efforts pursuing such paths. However, by determining that water vapor dramatically limits the energy concentration of collapsing bubbles, we thus have found a vector to lead us in new directions. We now work with this knowledge and search for low vapor-pressure fluids to enhance the collapse dynamics of bubbles.

Our progress with objective 3 has also been hampered, not by water vapor considerations, but by the difficulty of following the correct path towards increasing collapse dynamics with higher-order modes. We have found that a simple increase in a higher-order mode will not necessarily lead to a stable sonoluminescence regime. Furthermore, once at this new regime, we will still be confronted by the limitations imposed by vapor. Therefore, we believe that this line of approach, though useful in understanding bubble dynamics as a whole, will not lead us to new paths for dramatically increasing the collapse strength of bubbles. Therefore, we have limited this line of research.

The plasma state within the bubble was examined using x-rays. Unfortunately, no new information was obtained. The x-ray absorption into the water probably was too high for any appreciable energy to pass through the bubble. Furthermore, the plasma state within the bubble is not long-lived, leading to difficulties in timing the interaction between the x-ray beam and the bubble collapse. Due to the asynchronous relationship between bubble oscillations (near 20 kHz), and x-ray pulses (around 80 MHz), we seldom could conclusively determine if the picosecond time frame of the plasma state was being interrogated b y the x-ray beam. Until a synchronized system is used, it will be difficult and expensive to probe the bubble interior with x-rays.

We have achieved very interesting results with the lithotripter system. In Year 1, we discovered that the bubble collapse, more than 3 orders of magnitude greater than SBSL bubbles, did not produce any more light than an SBSL bubble. We used light scattering to measure the bubble motion (see Figure 3), and determined that water vapor trapping probably prevented the bubble from getting hot (Matula, Hilmo et al.).
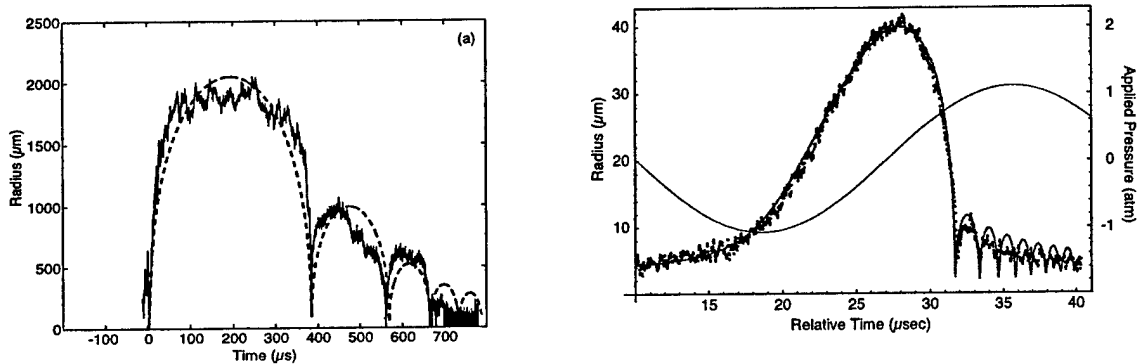
Figure 3. The left-hand-side shows measurements of bubble dynamics from a lithotripter pulse. The right-hand-side shows a typical SBSL bubble. Note that the SBSL bubble only grows to about 40-50 μm, while the lithotripter bubble grows to ≈2 mm. In both cases, the light emission intensity from the bubble collapse is about the same.

We then examined the light emission from the initial compression of preexisting bubbles and found that the light intensity was about 10 times greater than SBSL bubbles, even though the "starting" size of the bubble was only about 10 μm (see Figure 4). We interpret this to mean that the compression of a gas bubble generates much more heat than the collapse of a vapor bubble.
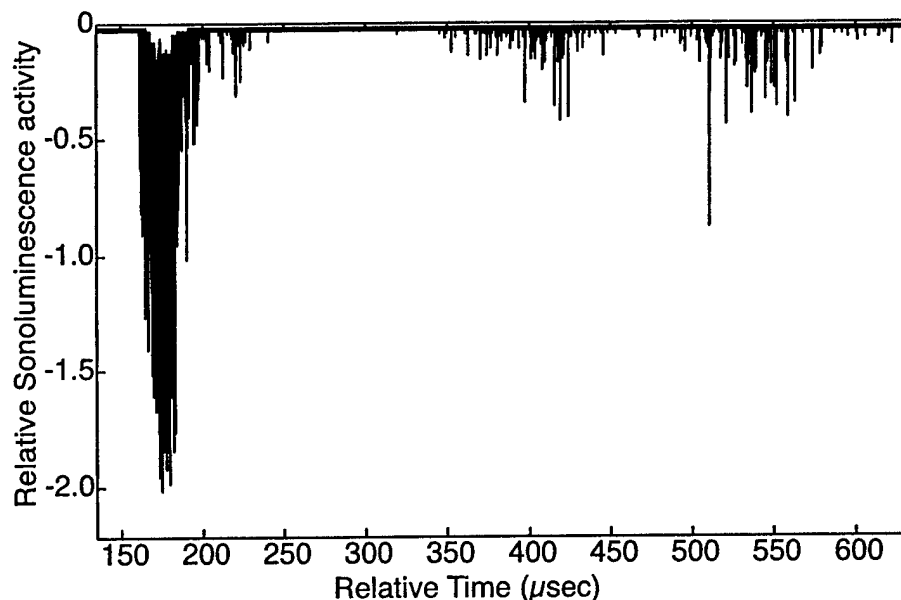
Figure 4*. Light-emission intensities from bubbles in a lithotripter. Approximately 1000 shocks were used, and the envelope is shown here. Notice that the maximum light emission intensity is at the beginning, during the compression phase of the bubble. There is only a small amount of light emission during collapse, and probably during the rebound, as well. For calibration, we used a normal SBSL system with the PMT located at the same geometrical distance as with the lithotripter. For SBSL, the light intensity is ≈ 0.25 V.*

We have recently set out to explore the effect of various fluids on lithotripsy pulses, and bubble collapse. Our initial results indicate that low vapor-pressure fluids do indeed generate much more light than high vapor pressure fluids. Our initial tests have been carried out with water, silicone oil, dodecane, and acetone. While dodecane and silicone oil generated much light, water, and especially acetone, generated very little light, as expected due to their high vapor pressure.

Holzfuss, J., M. Ruggeberg, et al. (1998). "Boosting sonoluminescence." Physical Review Letters 81(9): 1961-4.

Matula, T. J., P. R. Hilmo, et al. "Submitted for publication." Physics of Fluids.

Storey, B. D. and A. J. Szeri (2000). "Water vapour, sonoluminescence and sonochemistry." Proc. Roy. Soc. Lond. A 456: 1685-1709.

Toegel, R., B. Gompf, et al. (2000). "Does water vapor prevent upscaling sonoluminescence?" Physical Review Letters 85(15): 3165-3168.

OBJECTIVE

The objectives are (1) to build and test a shock-wave device for generating intense cavitation, and (2) to design, build, and test a high-pressure amplitude sine-wave cavitation generator. Our FY02 milestones included:
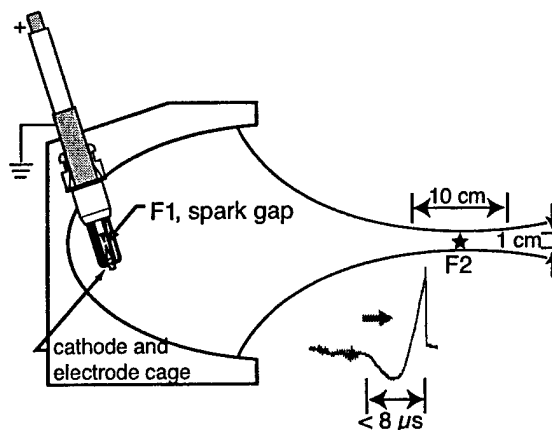
(1) To complete and test an electrohydraulic shock-wave device.
(2) To verify that the shock-wave device generates cavitation only when seeded.
(3) To design, build, and test a device that generates 20 atm of negative pressure.
(4) To verify that the device operates and generates cavitation only when seeded..

The goals of this research are to provide vectors for enhancing the light output.

APPROACH

For objective (1), we used our knowledge of lithotripters to build a higher-output shock-wave device. Our older research lithotripter uses a 20-kV voltage discharge across a set of electrodes immersed in water to generate a shock wave. The wave is reflected by an ellipsoidal reflector and focused to a small volume (F2); peak pressure amplitudes of up to 1,000 atmospheres positive and 100 atmospheres negative can be achieved with this system. Figure 1 illustrates our spark-generated lithotripter and a typical measured spatial waveform from our lithotripter. Based on our research lithotripter, we build a similar device, but with the ability to generate 40 kV, twice the voltage of our current unit. We also were able to modify the design to decrease the overall size of the device.

Figure 1. A diagram of the electrohydraulic shock wave lithotripter (SWL). A spark discharge located at the first focus of an ellipsoidal reflector is reflected by the ellipsoid and focused at the second focus, F2. Note that there also exists a direct diverging wave, originating from the spark gap, and passing through F2. Although small, the effect of this wave can be observed. The spatial profile of a measured shock wave is also shown.

Because normal single-bubble sonoluminescence (SBSL) only utilizes approximately 1 atmosphere (above ambient), we expect that the bubble dynamics from a lithotripter pulse to be much more energetic than from SBSL bubbles, possibly leading to much more interesting states of matter within the compressed bubble interior. Besides water, we also studied bubble effects from silicone oil, dodecane, and acetone.

For objective (2), we finalized on an array design for generating sine-wave pressures with amplitudes in excess of 20 atmospheres. The design is illustrated in Fig. 2. A series of transducers are mounted in a wheel assembly (Fig. 2 right) that sits near the top of a water-filled cylindrical vessel. The transducers generate downward going wave pulses that are reflected by an aluminum parabolic reflector. The sound waves then converge at the center of the vessel. If the path length of the transducers are equal, then the total

amplitude of the pressure wave is simply the addition of the individual amplitudes of each pressure pulse.
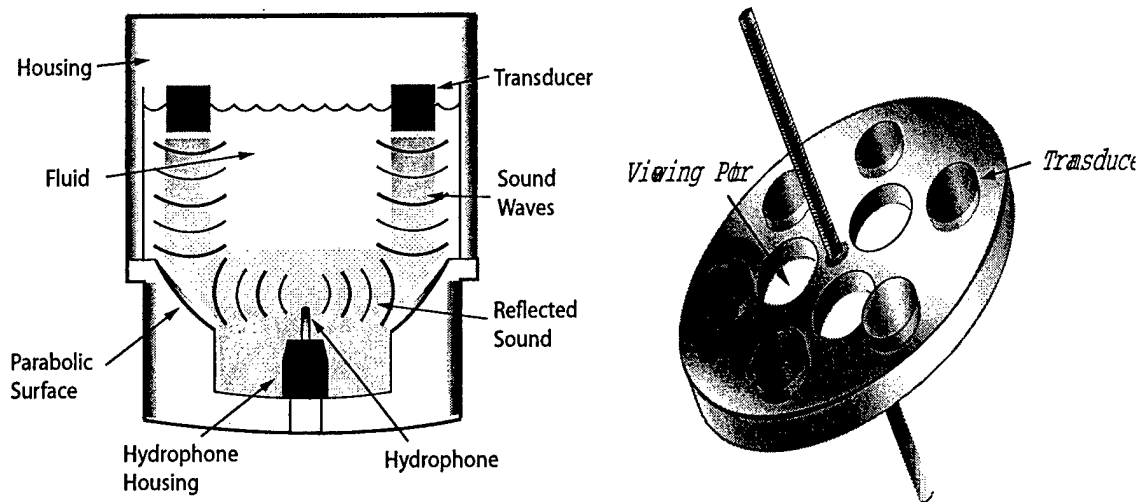
Figure 2. *An illustration of the sine-wave generator. Left: A transducer array sits partially immersed in a column of liquid. Wave trains travel downward until reflected by a parabolic reflector, and focus at the center. Right: The array sits in a wheel what is height-adjustable.*

PROGRESS

We have achieved very interesting results with the shock-wave system. We have made discoveries that have proven effective in focusing our work towards generating extremely intense bubble collapses. We discovered that the bubble collapse, more than 3 orders of magnitude greater than SBSL bubbles, did not produce any more light than an SBSL bubble. We used light scattering to measure the bubble motion (see Figure 2), and determined that water vapor trapping probably prevented the bubble from getting hot.
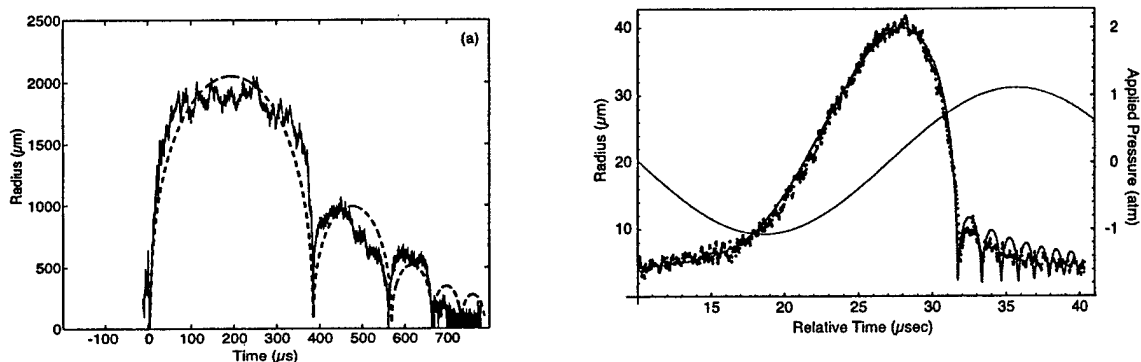
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We then examined the light emission from the initial compression of preexisting bubbles and found that the light intensity was about 10 times greater than SBSL bubbles, even though the "starting" size of the bubble was only about 10 μm (see Figure 3). We interpret this to mean that the compression of a gas bubble generates much more heat than the collapse of a vapor bubble.

Figure 3. Light-emission intensities from bubbles in a lithotripter. Approximately 1000 shocks were used, and the envelope is shown here. Notice that the maximum light emission intensity is at the beginning, during the compression phase of the bubble. There is only a small amount of light emission during collapse, and probably during the rebound, as well. For calibration, we used a normal SBSL system with the PMT located at the same geometrical distance as with the lithotripter. For SBSL, the light intensity is ≈ 0.25 V.
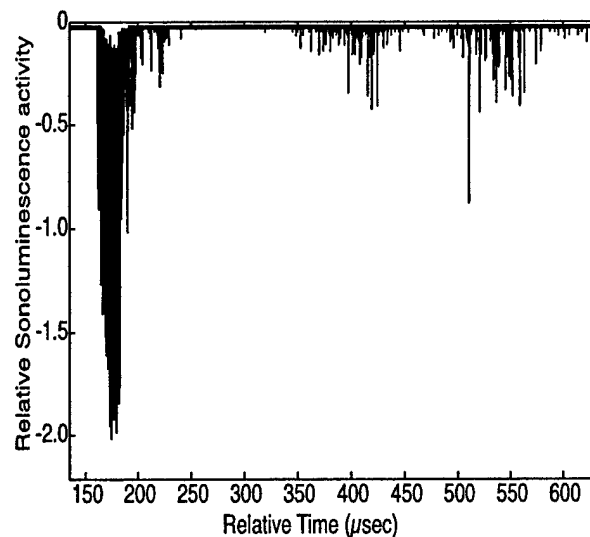
We explored the effect of various fluids on lithotripsy pulses, and bubble collapse. Our initial results indicate that low vapor-pressure fluids do indeed generate much more light than high vapor pressure fluids. Our initial tests have been carried out with water, silicone oil, dodecane, and acetone. While dodecane and silicone oil generated much light, water, and especially acetone, generated very little light, as expected due to their high vapor pressure.

Our most recent progress for this system involves the cooling and degassing of acetone for nucleation experiments. We have been able to cool the apparatus to 4°C, and with careful degassing, we have been able to remove most of the gas from the acetone. We are in the process of optimizing these controllable parameters.

Progress on the sine-wave array system has been impressive. We have completed the initial device, although optimization is still needed. Figure 5 illustrates the pressure and sonoluminescence generated at the focus. We used a calibrated B&K hydrophone to measure the pressure; only the negative portion is shown in this figure. Notice that sonoluminescence (and hence, cavitation) is initiated when the pressure exceeds about 20 atmospheres, although once cavitation is seeded, sonoluminescence can be obtained at lower pressures.

Figure 5. The negative pressure from a pulse of approximately 10 acoustic cycles is shown here. Note that the pressure reaches −20 atmospheres, as determined by a calibrated B&K hydrophone. The sonoluminescence from cavitation generated at these high pressures is also shown.
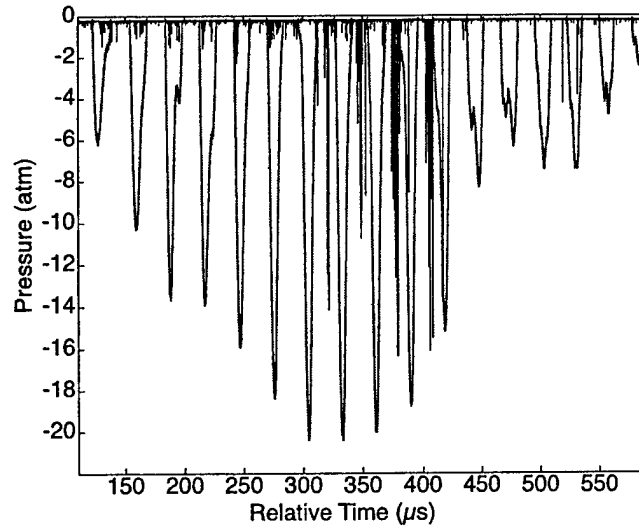
Current work for this system involves improving the transducer array; currently we use very simple and inexpensive transducers bought off-the-shelf. We plan to incorporate focusing lenses to increase the pressure amplitude. We are also working on methods to keep the fluid cold and degassed.